様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科研費 KAKENHI

科学研究費助成事業　　研究成果報告書

令和　４年　６月１４日現在

機関番号：８２４０１
研究種目：研究活動スタート支援
研究期間：2019～2021
課題番号：１９Ｋ２３５９３

研究課題名（和文）Exciton physics in 1D-2D heterostructures

研究課題名（英文）Exciton physics in 1D-2D heterostructures

研究代表者
Ｆａｎｇ　Ｎａｎ（FANG, NAN）

国立研究開発法人理化学研究所・開拓研究本部・基礎科学特別研究員

研究者番号：５０８５０５０９

交付決定額（研究期間全体）：（直接経費）　　2,200,000円

研究成果の概要（和文）：二次元材料はユニークな電子・光物性を持ち、次世代デバイスの材料として注目を集めています。六方晶窒化ホウ素（h-BN）は絶縁性の二次元材料で、不純物および欠陥が少なく、他の二次元材料の性能を引き出せることが分かっています。カーボンナノチューブは一次元材料であり、二次元材料同様に基板や表面の状態に敏感です。合成直後の架橋カーボンナノチューブは清浄な状態であり、光学特性は優れているのですが、シリコンなどの基板と接触すると消光効果が起き、 フォトルミネッセンスの強度は激減します。本研究では、h-BNとカーボンナノチューブのヘテロ構造を作り、h-BNとカーボンナノチューブの相互作用を調べました。

研究成果の学術的意義や社会的意義
These findings indicate that h-BN is an ideal solid-state subsatrate for the CNTs photonic devices and open a new path-way for manipulating excitons in CNTs.Moreover, we also observe that intimate contact with the h-BN substrate could result in large modifications in excitonic energies.

研究成果の概要（英文）：Hexagonal boron nitride (h-BN), a two-dimensional (2D) material, is atomically flat with low defect density, which is widely used to support other 2D materials for both electronics and photonics. We expect that the advantages of h-BN can also be utilized in mixed dimensional heterostructures, and carbon nanotubes (CNTs) would provide a unique opportunity in this context. The one-dimensional nature of CNTs results in enhanced Coulomb interactions, giving rise to tightly bound excitons that show photoluminescence (PL) at room temperature. CNTs directly attached on solid-state substrates such as SiO2/Si, however, suffers from the strong substrate quenching effect, hindering applications in all-solid-state optical devices. By using h-BN as a substrate, the quenching effect is expected to be suppressed. Moreover, excitons in CNTs are sensitive to the dielectric environment, and intimate contact with the 2D h-BN substrate could result in large modifications in excitonic energies.

研究分野：工学

キーワード：carbon nanotube　hexagonal boron nitride　exciton　screening effect

１．研究開始当初の背景

Hexagonal boron nitride (*h*-BN), a two-dimensional (2D) material, is atomically flat with low defect density, which is widely used to support other 2D materials for both electronics and photonics. We expect that the advantages of *h*-BN can also be utilized in mixed dimensional heterostructures, and single-walled carbon nanotubes (CNTs) would provide a unique opportunity in this context. The one-dimensional nature of CNTs results in enhanced Coulomb interactions, giving rise to tightly bound excitons that show photoluminescence (PL) at room temperature. CNTs directly attached on solid-state substrates such as SiO_2/Si, however, suffers from the strong substrate quenching effect, hindering applications in all-solid-state optical devices. By using *h*-BN as a substrate, the quenching effect is expected to be suppressed. Moreover, excitons in CNTs are sensitive to the dielectric environment, and intimate contact with the 2D *h*-BN substrate could result in large modifications in excitonic energies.

２．研究の目的

Here we investigate two different types of heterostructures consisting of *h*-BN and CNT by using PL excitation (PLE) spectroscopy. In samples where *h*-BN flakes are transferred on individual air-suspended CNTs, the chiralities are unambiguously assigned and the same tubes are tracked for *h*-BN effects. Bright luminescence with a narrow linewidth is observed from the CNTs directly attached to *h*-BN at room temperature, indicating weak quenching and small broadening effect from *h*-BN. Excitonic energies are found to exhibit considerable redshifts as well. The results demonstrate the ideal properties of *h*-BN as a substrate for CNT photonic devices

３．研究の方法

We begin by studying *h*-BN/CNT heterostructures consisting of an *h*-BN flake on top of an individual air-suspended CNT. First, the air-suspended CNTs are prepared on silicon substrates. Next, an *h*-BN flake is transferred on top of a CNT with the help of the marks on the substrate using a micromanipulator system. The *h*-BN/CNT structures are investigated with a home-built confocal PL microscopy system at room temperature. We characterize the *h*-BN effects on the CNTs by comparing PLE maps of individual CNTs before and after the transfer of *h*-BN. A clean PLE map can be obtained even after the transfer, since the PL intensity remains sufficiently strong. The reduction ratio of PL intensity after transfer compared to before transfer averages to 0.48 over 8 tubes, while the PL reduction ratio is typically less than 0.01 in the current solid-state substrates such as SiO_2/Si, quartz, and metals. The modest reduction of PL after the transfer of *h*-BN indicates the quenching effect from *h*-BN is weak, making *h*-BN a promising substrate for light-emitting CNT devices.
We point out that the full width at half maximum for E_{11} emission shows only a slight increase by 2.2 meV. The linewidth of *h*-BN/CNT is much smaller than CNTs on other solid-state substrates, while the inhomogeneous linewidth broadening of the ground exciton of WS_2 (WSe_2) in 2D WS_2 (WSe_2)/*h*-BN heterostructures is reported to be ~2 meV at low temperatures. The small change in the linewidth at room temperature suggest that our 1D/2D heterostructures show comparable interfacial properties to 2D/2D heterostructures.
The PLE maps also show substantial modifications of the excitonic energies. We identify the nanotube chirality from the PLE map before transfer to be (10,8) by extracting the E_{11} and the E_{22} energies from the emission and the excitation resonances, respectively. After the transfer of *h*-BN, we observe redshifts of 21 and 30 meV for E_{11} and E_{22}, respectively. We measured 18 more samples with different chiralities, and all the samples show redshifts for both E_{11} and E_{22}. Dielectric screening from *h*-BN is expected to cause some redshift, but the observed shifts are large considering the fact that *h*-BN is only attached to one side of the CNT.

４．研究成果

In summary, we have investigated *h*-BN effects on the optical properties of CNTs by performing PL spectroscopy on *h*-BN/CNT and CNT/*h*-BN heterostructures at room temperature. We have demonstrated that CNTs directly attached to *h*-BN are highly luminescent with narrow linewidths of ~12 meV, which is comparable to air-suspended

CNTs. The substrate quenching and broadening effects on the h-BN substrates are found to be much weaker than those in conventional substrates such as quartz and SiO_2/Si. In addition, the anomalously large redshifts in E_{11} and E_{22} are observed despite the fact that h-BN has a low dielectric constant and is only attached to one side of the CNT. These findings highlight the superior properties of h-BN for 1D/2D hybrid-dimensional photonics and open a new pathway for manipulating excitons in CNTs

５．主な発表論文等

〔雑誌論文〕　計2件（うち査読付論文　2件／うち国際共著　0件／うちオープンアクセス　0件）

１．著者名 N. Fang, K. Otsuka, A. Ishii, T. Taniguchi, K. Watanabe, K. Nagashio, and Y. K. Kato	４．巻 7
２．論文標題 Hexagonal Boron Nitride As an Ideal Substrate for Carbon Nanotube Photonics	５．発行年 2020年
３．雑誌名 ACS Photonics	６．最初と最後の頁 1773-1779
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acsphotonics.0c00406	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

１．著者名 Otsuka Keigo、Fang Nan、Yamashita Daiki、Taniguchi Takashi、Watanabe Kenji、Kato Yuichiro K.	４．巻 12
２．論文標題 Deterministic transfer of optical-quality carbon nanotubes for atomically defined technology	５．発行年 2021年
３．雑誌名 Nature Communications	６．最初と最後の頁 1-8
掲載論文のDOI（デジタルオブジェクト識別子） 10.1038/s41467-021-23413-4	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

〔学会発表〕　計5件（うち招待講演　0件／うち国際学会　1件）

１．発表者名 N. Fang, K. Otsuka, T. Taniguchi, K. Watanabe, K. Nagashio, Y. K. Kato,
２．発表標題 Dielectric screening effects on photoluminescence of carbon nanotubes on hexagonal boron nitride
３．学会等名 第67回応用物理学会春季学術講演会
４．発表年 2020年

１．発表者名 N. Fang, K. Otsuka, T. Taniguchi, K. Watanabe, K. Nagashio, Y. K. Kato,
２．発表標題 Dielectric screening effects on photoluminescence of carbon nanotubes on hexagonal boron nitride
３．学会等名 The 58th Fullerenes-Nanotubes-Graphene General Symposium
４．発表年 2020年

１．発表者名 N. Fang, K. Otsuka, A. Ishii, T. Taniguchi, K. Watanabe, K. Nagashio, Y. K. Kato,
２．発表標題 Hexagonal boron nitride as an ideal substrate for carbon nanotube photonics
３．学会等名 8th Workshop on Nanotube Optics and Nanospectroscopy（国際学会）
４．発表年 2020年

１．発表者名 N. Fang, K. Otsuka, T. Taniguchi, K. Watanabe, K. Nagashio, Y. K. Kato,
２．発表標題 Dielectric screening effects on photoluminescence of carbon nanotubes on hexagonal boron nitride
３．学会等名 PKU-Tsinghua-UTokyo Workshop on Nano Research
４．発表年 2020年

１．発表者名 Fang Nan
２．発表標題 Excitons in mixed-dimensional heterostructures
３．学会等名 「物質階層原理研究」&「ヘテロ界面研究」 2021年冬合同研究会
４．発表年 2021年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

—

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関